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Our goals were to investigate coherent effects, carrier dynamics and Robi Flopping, in semiconductors and study the nonlinear optics of semiconductor quantum dots. We been able to successfully complete these goals.

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Femtosecond Nonlinear Optics of Semiconductors
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N. Peyghambarian and S. Koch

Statement of the Problem Studied

Our goals were to investigate coherent effects, carrier dynamics and Robi Flopping, in semiconductors and study the nonlinear optics of semiconductor quantum dots. We been able to successfully complete these goals and produce 357 publications and presentations as outlined below.

Summary of Results

1. Ultrafast transient gain in type II multiple quantum wells

Subpicosecond gain was found in type II multiple quantum wells and compared to the gain dynamics in type I multiple quantum wells. For the measurements a new experimental technique was developed which enables the observation of gain decay into absorption with previously unachieved precision and which should also be of relevance in other similar experiments.

We used a three beam setup for the investigation: a first pulse (gain pulse) was used to optically inject a high carrier density into the sample and create gain with the following two pulses a pump probe experiment was performed in the gain region of the inverted semiconductor. The second pulse (pump pulse) in this case causes stimulated recombination, depleting the carrier distribution around the pump frequency and burning a spectral hole. This leads to induced absorption at the pump wavelength which is subsequently tested with the third, broad band pulse (probe pulse). The depth of the spectral hole is now proportional to the amount of gain present in the sample before the pump pulse arrived. Keeping all other experimental parameters constant, especially the pump intensity and the pump-probe delay, it is possible to map out the development gain and its decay into absorption by monitoring the depth of the spectral hole as a function of gain delay.

This procedure was carried out on two samples, both grown by molecular beam epitaxy. The first sample consisted of 150 periods of 28 A GaAs wells separated by 56 A AlAs barriers, resulting in type II electronic confinement. The second sample was made up out of 150 periods of 26 A GaAs wells and 80 A AlGaAs barriers with an Al content of 41% yielding type I confinement. Both samples were kept at low temperatures. Figure 1 shows our experimental findings. It shows the temporal behavior of the spectral hole in type II (part (a) of the figure) and the type I (part (b) of the figure sample.

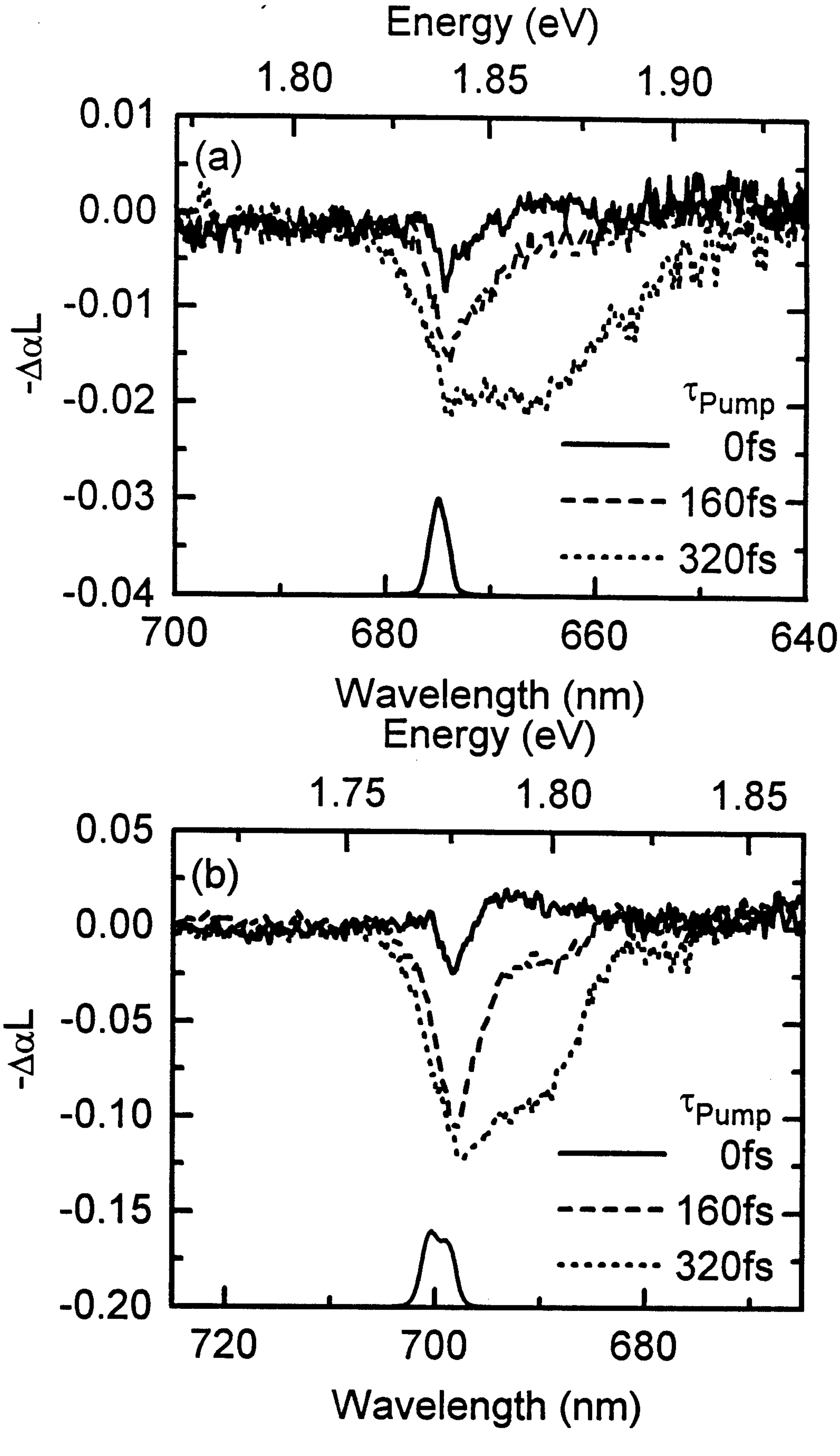

Figure 1. Spectral hole dynamics in the type II (Fig. 2(a)) and type I (Fig. 2(b)) sample at low temperature. The curves are 160 fs apart and gain delay is fixed at 500 fs. The pump spectrum is also shown in the lower portion of each graph.

The experimental findings can be explained by the different scattering dynamics of the electrons in the two samples. Figure 2 shows a schematic of the electronic energy levels. The lowest confined conduction band state lies within the barrier of the type II multiple quantum well at the X-point of the Brillouin zone. Optical absorption, however, takes place at the Ã-point of the well. Carriers created at the Ã-point scatter to the lower lying state in the barrier resulting in a separation of electrons and holes in real and momentum space. This separation inhibits stimulated recombination between the carriers and therefore gain. Since the scattering time is very short for this process, the carrier population at the Ã-point decays very rapidly and with it the inversion and gain. In the type I sample on the other hand, the lowest lying electronic state is at the Ã-point and the carriers remain there until they recombine. Here the gain decay is therefore governed by the carrier lifetime, which is much longer than the time constant for the previously described scattering process in the type II sample.

2. Observation of charge carrier LO-phonon emission in cold plasmas

Spectral hole burning measurements were performed in the gain region of the type I sample described above at very high carrier densities. The experimental setup employed was similar to that used in the transient gain measurements. Figure 3 shows the results. Besides the spectral hole around 1.775 eV a second structure can be found at approximately 40 meV higher energy. A third peak can also be observed at approximately 80 meV higher energy than the original spectral hole. These replicas of the spectral hole are explained by LO-phonon emission of higher energy carriers, that scatter into the vacancies of the spectral hole. A simple model based on the Boltzmann equation for carrier LO-phonon scattering in connection with the noninteracting version of the semiconductor Bloch equations for two bands was developed to describe the experimental findings. The results are shown in Fig. 5. The same replicas of the spectral hole as in the experiment were found. Carrier-carrier scattering has been neglected in the calculations for clarity.

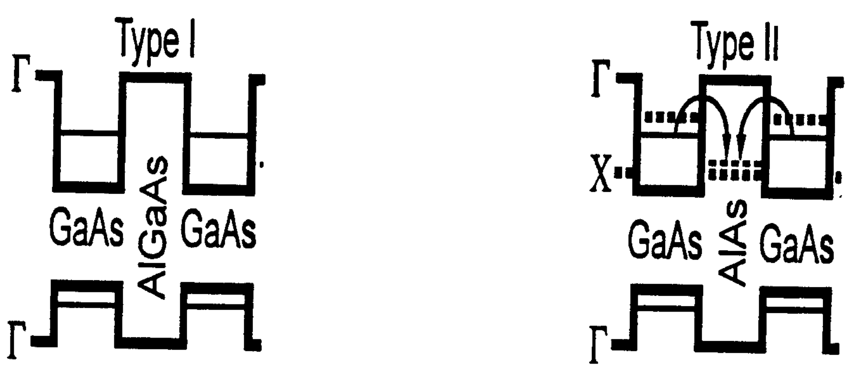

Figure 2. Energy level diagram for the type II and the type I sample. The thick solid lines represent the bandgap energy at the Ã-point whereas the thick dashed lines the bandgap energy at the X-point of the Brillouin zone indicate. Thin dashed lines stand for the lowest quantum confined states. The arrows indicate the scattering process in the type II sample.

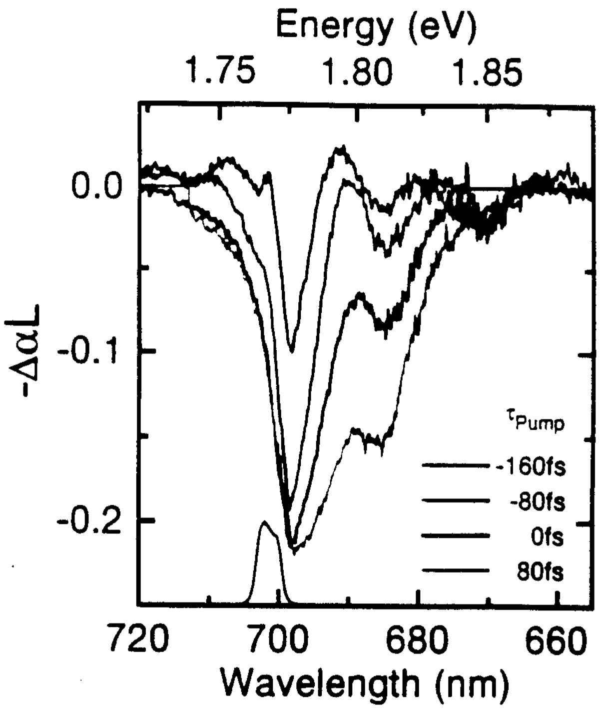

Figure 3. Experimental spectral hole burning results for 1, 10 and 20 ps gain delay. Shown is the differential absorption vs. wavelength for four different pump-probe delays. A spectral hole is clearly visible around 1.77 eV at all three gain delays. Additional peaks can also be observed for 10 ps and 20 ps gain delay.

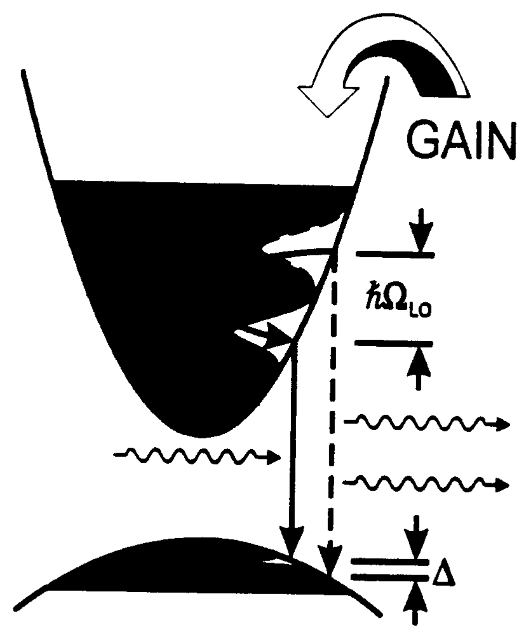

Figure 4. Schematic for development of a spectral hole replica. Higher energy electrons scatter into the vacancies of the original spectral hole, creating a similar nonequilibrium distribution higher in the band than that was burned by the pump pulse.

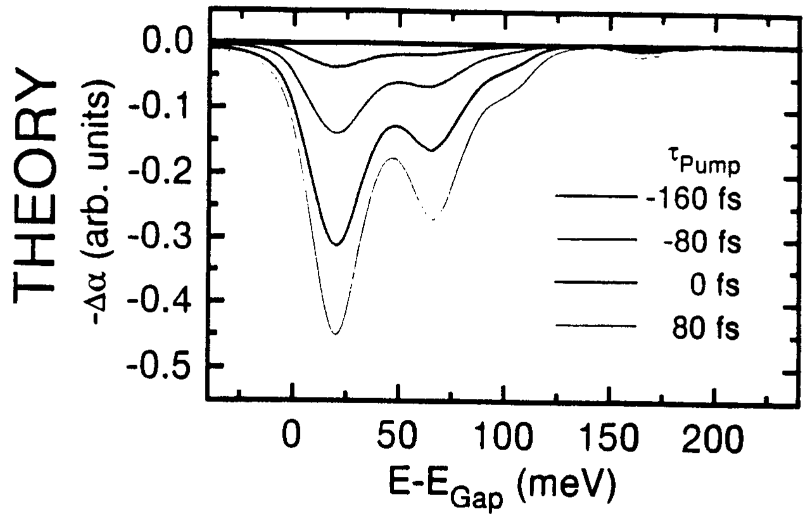

Figure 5. Calculated spectral hole replica.

3. Spectral hole burning in the gain region of an inverted semiconductor

We obtained the first conclusive evidence for spectral hole burning in the gain region, studied the gain dynamics, and compared our experimental results with calculations that include the full many-body effects. We obtained an excellent agreement between the theory and experiment. In the experiments, we used three separate femtosecond pulses (using our CPM system), consisting of a "gain" beam which excited the sample and created the gain, a tunable pump beam that depletes the carriers in the gain region, and a broad band probe to observe the effects of the pump beam. Figure 6(a) shows the temporal development of a spectral hole while pumping a CdSe thin film at 707 nm. The earliest curve shows a definite spectral hole developing around the pump.

Our theoretical analysis was based on the semiconductor Bloch equations and contained the Coulomb potential acting between the charge carriers, which in turn is screened by the carriers. These equations can be derived from nonequilibrium Green's functions as well as other approaches.

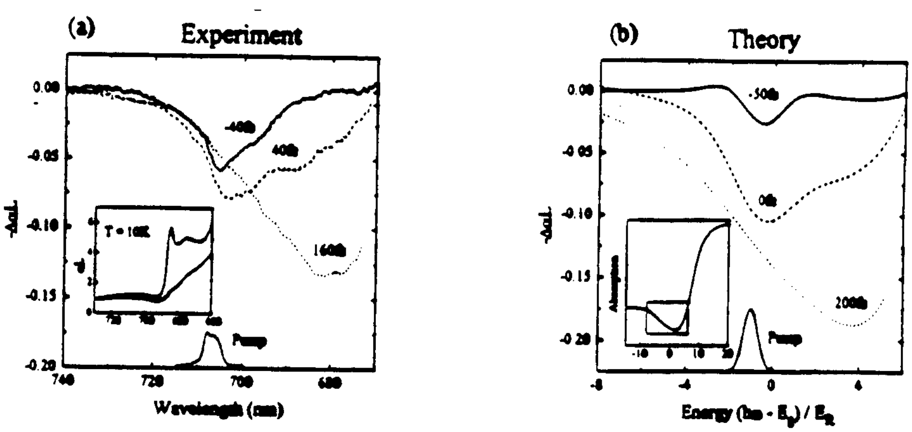

Figure 6. (a) Measured dynamics of the spectral hole with a pump beam of 707 nm. The inset shows the absorption with and without the gain beam. (b) Calculated probe differential absorption spectra for bulk CdSe at three pump-probe delay times.

Results of our theory are presented in Fig. 6(b) and good agreement between experiment and theory can be seen by comparing Fig. 6(a) and 6(b).

4. Carrier dephasing in the gain region of an inverted semiconductor[2,6-8]

We investigated the carrier dephasing well below and above the transparency point (TP, or the quasi-chemical potential) in a dense plasma prepared by optical pumping of a semiconductor at low temperatures. Optical excitation by a strong femtosecond pulse situated far above the band edge prepared a large spectral region of optical gain extending many meV below the transparency point. To measure the polarization dephasing rate in this strongly inhomogeneously broadened region, both spectral hole burning and time-integrated four-wave mixing (TI-FWM) were used. Figure 7 shows the results of the T_2 measured spectrum and its comparison to the theory. Good agreement is obtained.

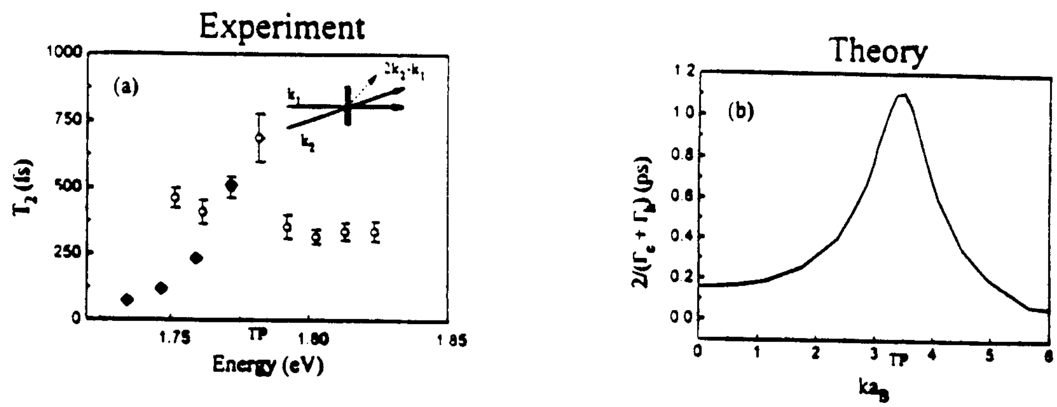

Figure 7. (a) T_2 times obtained from time-integrated self-diffraction (circles with error bars and from spectral hole burning). (b) Calculated dephasing times from carrier-carrier scattering.

5. Rabi oscillations in semiconductors

We theoretically and experimentally investigated the case of resonant exciton excitation where many-body effects play a significant role in the ultrafast response of the semiconductor. It should be recalled that in the case of resonant excitation in discrete atomic systems, the dipole-coupled level populations undergo Rabi oscillations. We found that when excited at or above the exciton resonance, the semiconductor carrier density clearly exhibits temporal oscillations. Moreover, the number of Rabi flops is basically twice that expected for a two-level system. A careful analysis shows that the Coulomb exchange effect renormalizes the field, and thus, accounts for the doubling of the Rabi flops under the assumed conditions.

The experimental verification of these calculations were published in our publication number 14.

6. Transient absorption of ultrafast pulses in an inverted semiconductor and electric field effects

We obtained experimental and theoretical evidence for a new effect: transition from amplification to absorption for very strong femtosecond optical pulses centered in the gain spectrum of a semiconductor laser diode amplifier. We predicted that a high-intensity femtosecond pulse will actually be absorbed by a semiconductor gain medium even when the pulse frequency spectrum is completely within the gain bandwidth. The induced absorption is identified with an interaction of the laser pulse with the tails of the high-lying non-inverted continuum states.

We performed experimental transmission, cross-correlation, and spectral measurements of 135-fs FWHM pulses propagating through a semiconductor laser diode amplifier prepared with antireflection coated facets. We tuned a self-modelocked Ti:sapphire laser operating at 90 MHz across the bandedge of a Hitachi HLP1400 laser diode with 30 mA of injection current. With the laser tuned to the peak of the gain, the laser diode initially exhibited gain saturation as the input pulse energy was increased, but rather than asymptotically approaching unity gain, the output pulse energy continued to decrease as the pulse underwent induced absorption.

We also investigated generalized Block oscillations and related electric field effects in semiconductor heterostructures.

7. Quantum Dot Gain and Lasing

We investigated the ultrafast optical dynamics of semiconductor quantum dots in the strong quantum confinement regime, where the bulk excitonic Bohr radius a_B is larger than the dot radius. We chose InP quantum dots in a toluene solution with radii of 25 and 35 Å (a_B = 100 Å), and CdSe quantum dots in a borosilicate glass matrix with an average radius of 25 Å (a_B = 58Å).

We were able to demonstrate quantum confined states at room temperature in InP samples for the first time. Femtosecond spectral hole burning spectroscopy revealed the dynamics of the lowest lying state: after a bleaching rise time of a few hundred femtoseconds, which follows the pump pulse of 115 fs, a bleaching recovery time of more than 200 ps was measured. We concluded that the carriers created by the femtosecond pump beam were trapped in long living trap states. Therefore, we concluded that we could resolve the broad homogeneous linewidth within the inhomogeneous broadening that is due to size distribution of the dots.

The CdSe quantum dots in the glass matrix were investigated at 10 K, and for the first time, it was possible to demonstrate gain in strongly confined zero-dimensional systems (see Fig. 8). From the gain buildup and decay dynamics (see Fig. 9), we were able to come up with a theoretical model including excitons and biexcitons to describe the gain behavior. A theoretical model that used a matrix diagonalization method to calculate the one-pair and two-pair states and used the optical Bloch equations to describe the multilevel dynamics agreed very well with our observations. The most important conclusion from our measurements was the fact that in any quasi zero-dimensional system, the gain is not narrow, as was often assumed, but quite broad and can reach a width of 100 nm. The gain stretches below the absorption edge, not due to bandgap renormalization, but due to biexciton to excited exciton transitions. Femtosecond spectral hole burning spectroscopy in the gain region confirmed the proposed model and showed that spectral holes in the gain region are replenished within a few hundred femtoseconds. This is an important aspect for possible device applications. The broad gain region could lead to short pulses if some modelocking were introduced.

Low density spectral hole burning measurements in the CdSe quantum dots showed no signs of a phonon bottleneck; only very fast carrier relaxation times of below one picosecond could be found. Because of the fact that the gain is due to biexciton to exciton transitions, the question of the existence and the implications of the phonon bottleneck are no longer important for device applications.

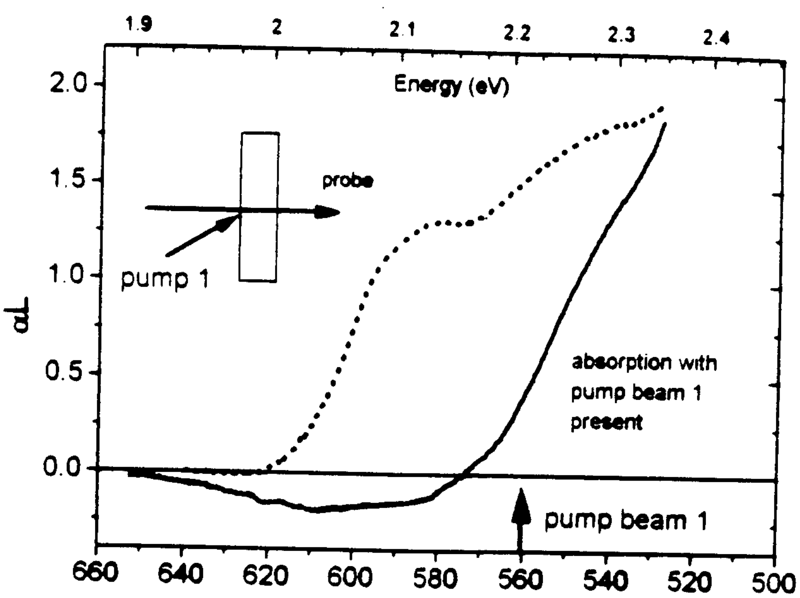

Figure 8. The dotted curve shows the linear absorption spectrum of the CdS quantum dot sample. The solid curve shows the absorption spectrum when the pump (at λ = 560 nm) is present. The pump beam creates carriers that change the absorption spectrum. A large region of gain (negative absorption) is seen. The gain extends from 650 nm to 572 nm. The gain in the region from 620 nm to 650 nm occurs where no absorption is present. This gain is associated with the "biexciton gain."

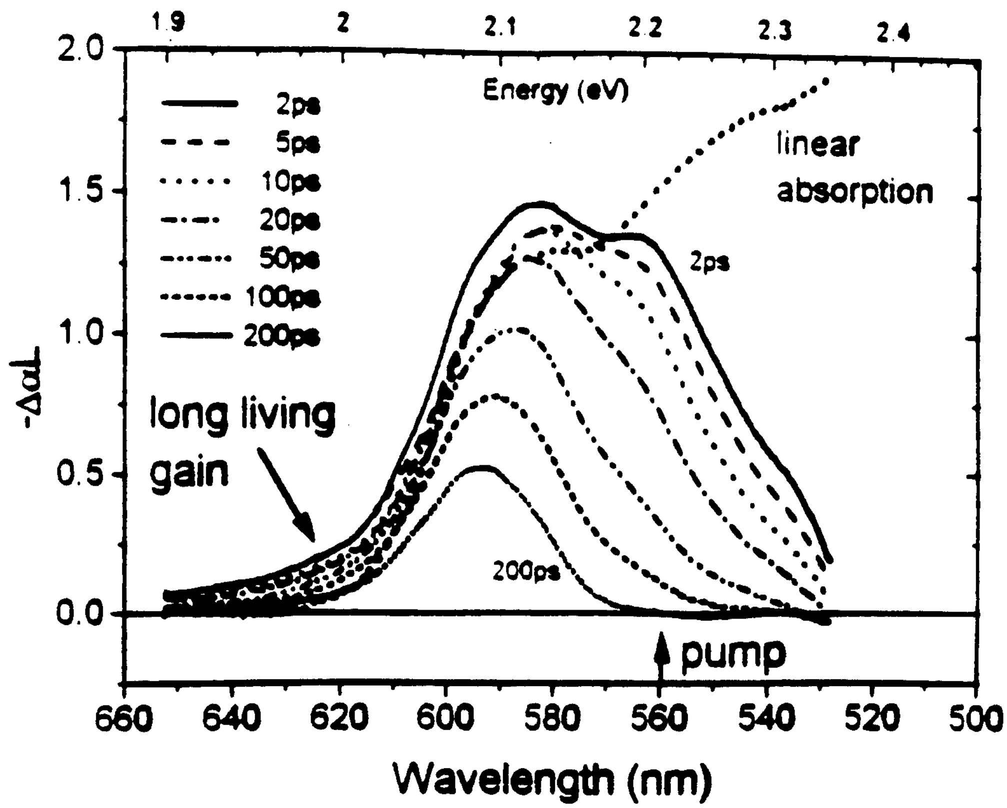

Figure 9. Gain dynamics measured with femtosecond laser pulses. The gain reaches its maximum in 2 ps. The gain decay occurs in a few hundred ps.

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